Cobalt-Free Cathodes for Next Generation Lithium-Ion Batteries

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Project ID: BAT417

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Overview

Timeline

- Project Start Date: 01/01/2019
- Project End Date: 12/31/2021
- Percent Complete: 10 %

Barrier and Technical Targets

- Barriers addressed:
  - Cycle Life: 1000 cycles C/3 deep discharge with < 20 % energy fade
  - Cost: < $100/kWh

Budget

- Total Project Funding: $3.08 M
  - DOE share: $2.46 M
- Funding for FY2019: $1.04 M
  - DOE share: $805k
- Funding for FY2020
  - DOE share: $818k

Partners

- Ohio State University: Jung-Hyun Kim
  - Battery testing
  - Cell chemistry development
- Navitas Systems Michael Wixom
  - Large-scale electrode fabrication
  - 2-Ahr battery manufacture and testing

Relevance

Impact

- Renewed interest in reduced or cobalt free Li-ion battery cathode formulations
- Innovate high-voltage Li-Ion batteries by producing an effective kinetic barrier that can act as a cathode SEI layer and enhance battery stability

Project Objectives

- Demonstrate lithium manganese nickel titanium oxide (LNMTO) as a high performance and cobalt-free Li-ion battery cathode material
  - Maintain high specific performance associated with LNMO
  - Enhance cycle life to achieve > 1000 cycles
  - Identify low-cost, scalable manufacturing process for LNMTO cathode powder

Objectives Budget Period FY19

- Down-select solid-state synthesis process for manufacture of high-voltage spinels
- Demonstrate applicability of LNMTO based cathodes through 2-Ahr testing

Approach/Strategy - Milestones

Milestone/Decision Point	FY 2019 Q1	FY 2019 Q2	FY 2019 Q3	FY 2019 Q4	FY 2020 Q1	FY 2020 Q2	FY 2020 Q3	FY 2020 Q4
1st iteration of solid-state LNMTO powder delivered	◆							
Decision-Point: Down-selection of solid-state synthesis process *Compositional homogeneity and coin-cell performance*		◆						
Down-selection of co-precipitation LNMO process				◆				
Decision Point: PPC chemistry, fabrication/conditioning process defined			◆					
2-Ahr PPCs fabricated and delivered to DOE *20 PPCs fabricated, 15 delivered to DOE | 15 completed performance testing*				◆				
Go/No-Go Decision Point: Performance testing of PPCs completed Achieve meaningful improvement over current LNMO, equivalent to LMNTO				◆				
PPC post-mortem analysis completed					◆			
Decision Points: Candidate core-shell LNMTO powders down-selected						◆		
Promising cell chemistries down-selected							◆	
Go/No-Go Decision Point: Testing of 2-Ahr cells completed *Demonstrate that core/shell modification enhances cathode performance*								◆

- Greater than 200 cycles with < 20 % energy fade (C/3, 25 °C)
- Specific energy > 650 Wh/g

- Cycle life > 500 cycles with < 20 % energy fade (C/3, 25 °C)
- Specific energy > 650 Wh/g

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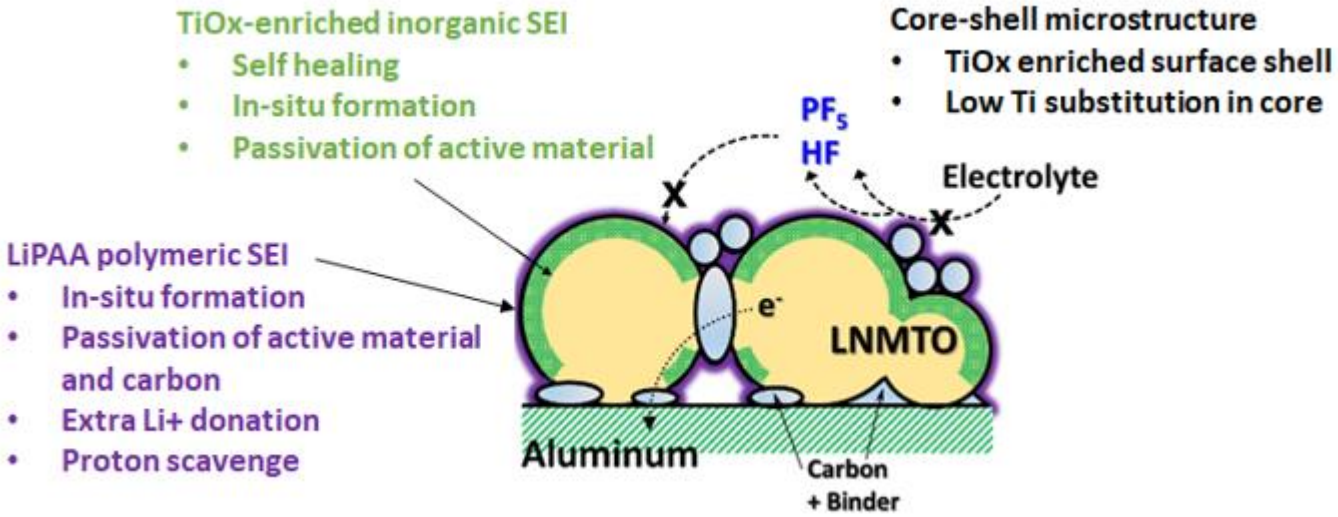
Approach

Develop cobalt-free cathode based on high-voltage $LiNi_{0.5}Mn_{1.5}O_4$ (LNMTO)

- Improve cycle and calendar life of high-voltage spinel cathodes by forming a solid-electrolyte interface that effectively passivates the cathode surface
- Microstructural enhancement of the LNMTO powder to create novel core/shell structures where titanium is preferentially located at the surface
- Incorporate optimized binder/electrolyte chemistries to address degradation



Technical Progress: Solid-State Processing

Successfully manufactured LNMTO cathode powder using solid-state synthesis

Synthesized high voltage spinel cathode materials using solid-state process

- Baseline LNMO
- Process iterations have shown steady performance improvement for LNMTO

Identified key process parameters

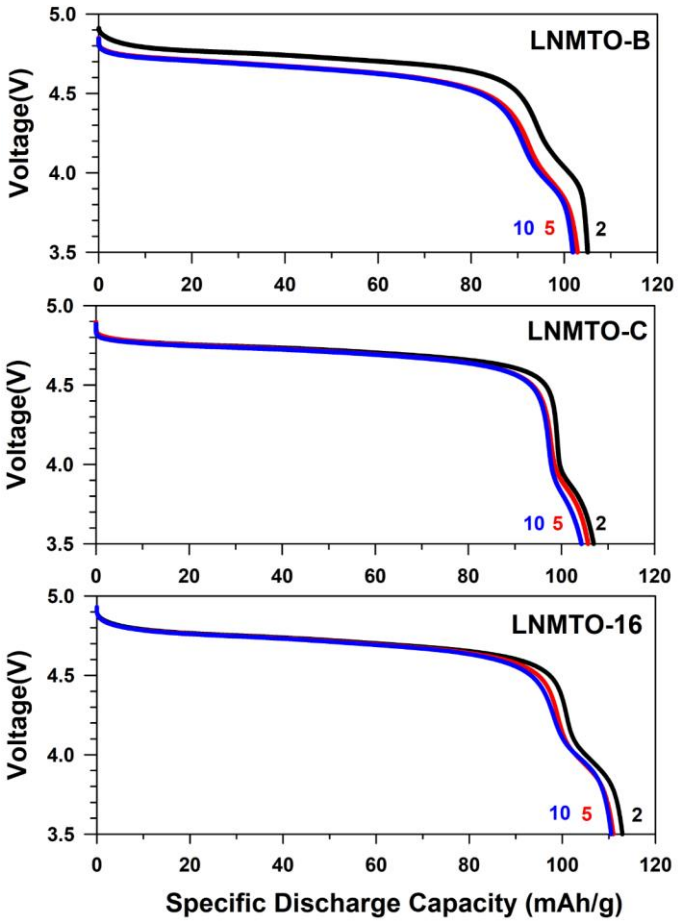
- Integrated new mixing process
- Optimized calcination profile
- Evaluated post-fire annealing process

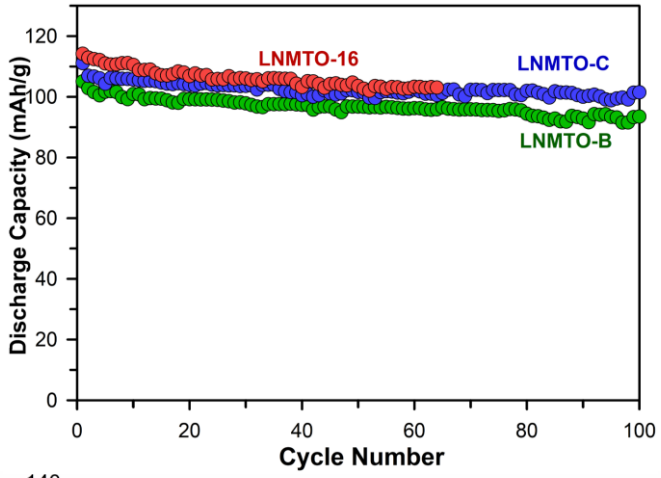
Established baseline manufacturing/cost model

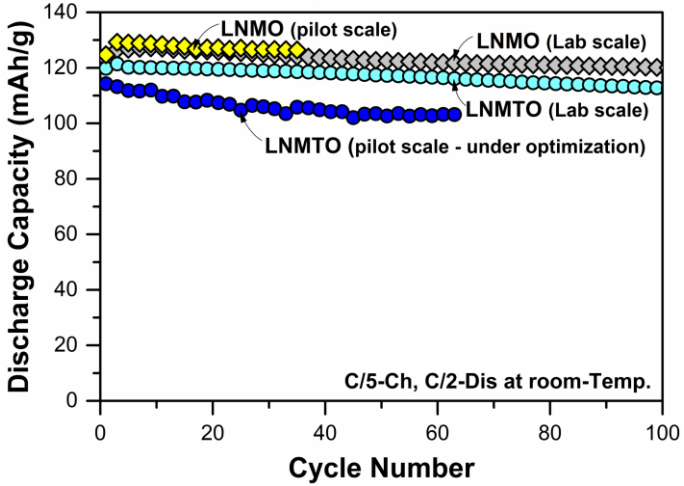
Technical Progress: Preliminary Cell Testing

Demonstrated cell performance of LNMO and LNMTO solid-state spinel powders

Process Iteration







Collaboration and Coordination with Other Institutions

Collaboration	Role
The Ohio State University	**Sub-contractor, University** ○ Coin-cell and single-layer pouch cell screening of cathode materials ○ Cell chemistry (additives/binder) development ○ Analytical characterization of cathode materials and electrodes
NAVITAS SYSTEMS	**Sub-contractor, Industry** ○ Electrode scale-up ○ Large format 2-Ahr battery fabrication and testing

Nexceris is working to identify additional opportunities to collaborate both within and outside of this project

- Partners for high volume manufacturing

Proposed Future Research (FY2019 and FY2020)

Ongoing Work FY 2019

Future Work	Justification	Key challenges
Complete down-selection of solid state LNMTO synthesis process and cell chemistry *Q2 and Q3 milestones*	○ Identification of optimal LNMTO powder & complimentary cell chemistry for PPCs	○ Complete process development in time ○ Risk mitigation for large 2-Ahr battery fabrication and testing
Fabrication & performance testing 2-Ahr cells *Q4 milestone and FY2019 Go/No-Go DP*	○ Demonstrate technology applicability ○ FY2019 deliverable	○ Compressed timeline ○ Coordination of testing protocol
Develop LNMO co-precipitation process *Q4 milestone*	○ Required for core/shell development in FY2020	○ Homogeneity of LNMO powder

Proposed Future Work FY 2020

Future Work	Justification	Key challenges
Post Mortem analysis of PPCs *Characterize surface of aged electrodes*	○ Identify opportunities to improve cell performance (cathode powder and/or cell chemistry)	○ Ensure primary degradation mechanisms are addressed, and mitigation strategies incorporated into work-plan
LNMTO core/shell microstructure development *LNMO core with TI-enriched surface*	○ Increase cycle-life w/o degrading capacity ○ Establish strong IP position ○ Pathway to achieve Y2 Go/No-Go DP	○ Efficient screening of candidate processes ○ Uniformity of Ti-enriched surface layer
Tailoring of cell chemistries for high V cathode *Stabilization of electrode/electrolyte interfaces*	○ Critical to implement to achieve required cell stability and cycle life	○ Identification and validation of cell chemistries for high V cathodes

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Summary

Accomplishments:

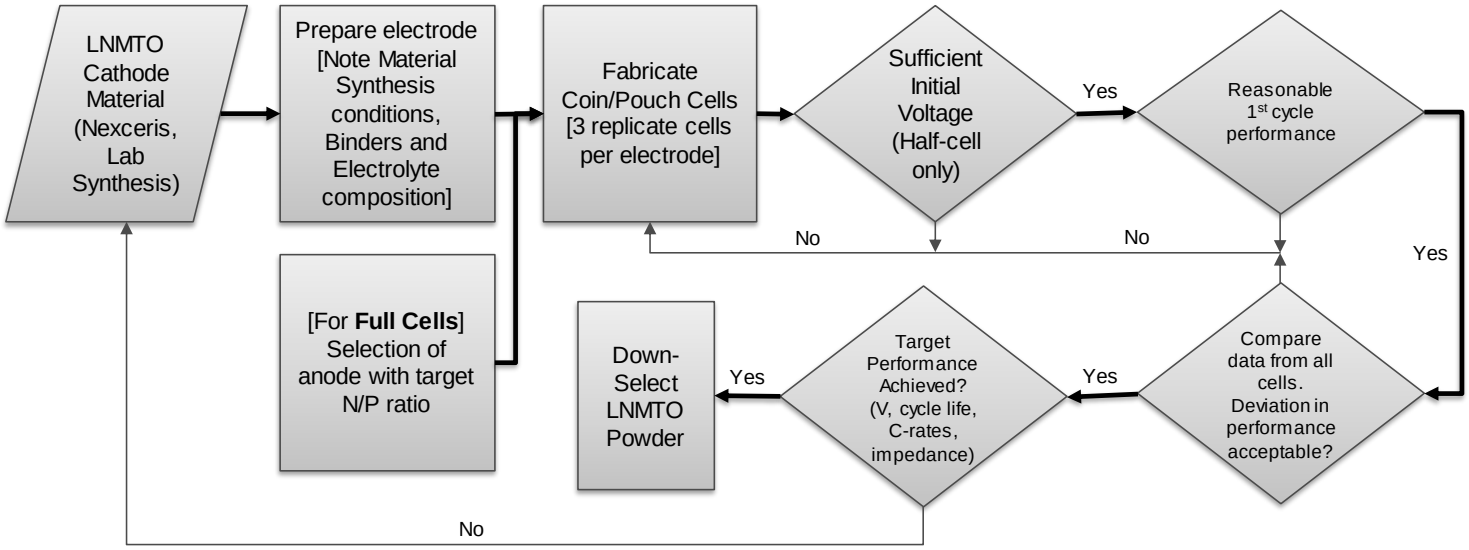
- Developed solid-state synthesis process for high-voltage spinel cathodes
- Solid-state LNMO and LMNTO powder successfully synthesized and tested
  - LNMO: Ti-free $LiNi_{0.5}Mn_{1.5}O_4$ (LNMO) has been successfully produced at pilot scale with ~ 130 mAh/g initial discharge capacity and 97.7% capacity retention @ 35^{th} cycle (C/5-Ch & C/2-Dis at RT)
  - LNMTO: Ti-substituted $LiNi_{0.5}Mn_{1.2}Ti_{0.3}O_4$ has been developing at pilot scale and showed performance improvement: from 105 to 115 mAh/g initial discharge capacity

Next steps:

- Complete down-selection of solid-state process and cell chemistry
- Risk mitigation and preparation for successful manufacture and testing of PPCs

Technical Back-Up Slides

Cathode Powder Screening Protocol



Battery Cell Testing (2032 stainless coin-cell)

- Positive Electrode: LNMTO active metal (85 wt.%) + Super P (7.5 wt.%) + PVdF (7.5 wt.%)
- Negative Electrode: Li-metal
- Separator: Polypropylene (Celgard 2500, 25 µm-thickness)
- Electrolyte: 1 M LiPF6 in 1:1 EC/EMC (Sigma-Aldrich)

Tested with Arbin LBT cycler at RT

Effect of post-process anneal

1-2 hour anneal improves LNMTO performance